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## ATOMS IN STRONG ELECTROMAGNETIC FIELDS

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I will take this opportunity to report on some fundamental calculations of atomic physics necessitated by having to take into account the fact that some current lasers field strengths in the focal region are comparable to or even greater than atomic fields. The time-honored technique of perturbation theory become prohibitively difficult and for sufficiently large intensities inadequate, and new techniques must be found to describe the laser-plasma interaction. Transitions involving the absorption or emission of many field quanta can occur under these circumstances.

However, since the laser frequency,  $\omega$ , is in general much less than atomic frequencies we are able to remain within the classical description of the electromagnetic field and are led to develop a description asymptotic in  $\omega/\Delta$  where  $\Delta \equiv (E_2 - E_1)/2$  and  $E_1$  and  $E_2$  are two characteristic atomic frequencies.

I shall comment briefly on three pieces of work : multiphoton excitation with a simple atomic model; multiphoton ionization again within a simple model; and finally a treatment of multiphoton ionization of a complex high  $Z$  atom in which we argue for a modified Thomas-Fermi description.

### 1) Multiphoton excitation - two level system

Most experimental studies involving multiphoton processes have investigated multiphoton ionization. There is the suggestion, however, based on Rhodes' observation<sup>1</sup> in Krypton, that excitation by a many photon process has been observed experimentally. In light of these results, it has been proposed that selective excitation by low-frequency electromagnetic fields may be feasible and desirable.

The simplest model that addresses this problem is the two-level system given in viewgraph 3. There  $E_2 > E_1$ ,  $\omega > 0$ ,  $F > 0$ ,  $\dot{F}/F \ll \omega$ . Here  $F$  is proportional the laser field strength and the dipole moment connecting the two states. The field is treated semiclassical and in the dipole approximation - laser wavelength much greater than atomic size. (This model describes other physical problems such as a spin -  $1/2$  particle in crossed static and oscillating magnetic fields.) Solutions of this problem have been obtained from perturbation theory ( $F_{\text{small}}$ ) by Shirley<sup>2</sup> using a Floquet analysis and perturbation theory. We develop a theory in  $\omega/\Delta$  small but spanning the domain  $F \sim \Delta$  and constrained only by  $F \ll \Delta^2/\omega$ . The identification with the quantized treatment is that  $2\Delta/\omega$  is taken equal to the number of photons involved in the transition.

We first discuss the case where  $F$  is constant. In order to find the time evolution of the system we write the first order system in second order form, viewgraph 4, and introduce the WKB representation. Solving the problem then amounts to find the small changes in the amplitudes of this representation over each period of the applied field. Exactly as in the problem of calculating the classically forbidden above barrier transmission coefficient<sup>3</sup> or, isomorphically, the change in the adiabatic invariant, the small changes necessitate moving our WKB path for the phase integral into the complex-time plane to meet the the, in this case, zeroes of  $Q$ , where the WKB breaks down. By asymptotic matching to the solutions in the vicinity of  $Q = 0$ , or equivalently using the established connection formulas for the jumps in the solution on crossing a Stokesline in one of these regions (viewgraph 5) we determine a transfer matrix which advances the system over one period of the applied field. The  $N$ th power of this matrix then determine the state of the system after  $N$  periods. The method is to be contrasted with Shirley's approach using Floquet theory, where it is necessary to find the eigenvalues of very large matrices.

We are thus able to find the excitation probability  $P$  of finding a particle in state 2 given that initially it was in state 1 in terms of the real phase advance per half period  $\theta_0$  and the phase integral  $\varepsilon \equiv (\tau_{k^-}, \tau_{k^+})$  between conjugate zeroes of  $Q$  (see viewgraphs 6, 7, 8 and 9) which quantities are functions of field frequency and intensity. For small  $F/\Delta$  we recover the resonance shifts and  $(F^2)^{\Delta/\omega}$  power for the generalized Rabi

frequency<sup>4</sup> as found by a perturbative treatment of Shirley's Floquet Hamiltonian. Simplified expressions for  $E$  and the resonant frequency,  $\omega_{\text{res}}$ , can also be given for  $F/\Delta \ll 1$ . We note the exponential smallness of  $u$  since  $\omega F/\Delta^2 \ll 1$ . Because of the smallness of  $u$  the resonance widths for  $P$  are very small. Further to the extent our model is applicable we see variations in  $\omega_{\text{res}}$  that are large compared to  $u$  will occur in practice because of intensity variation due to finite pulse length. These considerations place very stringent criteria on the constancy of the intensity if any particular resonance is to be maintained for as long as one Rabi period. In fact, because of the intensity dependence of  $\omega_{\text{res}}$ , the condition  $\omega_{\text{res}} - (2p+1)\omega = 0$  for a given order  $p$  is satisfied precisely at only a single time  $t_p$ . If  $p \gg 1$  then many resonances are passed over as the applied pulse evolves. (See viewgraph 10) In the short autocorrelation time limit in which successive resonances contribute with random phases, the average excitation rate is given by  $R = \pi u^2/4\omega$ .

To test the relevance of this result to proposed realistic experimental conditions<sup>5</sup>, this average excitation rate was evaluated for parameters relevant to the case of multiphoton excitation in  $\text{Cd}^{12+}$ , and was compared to the rate of multiphoton ionization from the upper excited state using the formula of Keldysh<sup>6</sup> ( $\gamma \ll 1$ ). The ionization from the excited state was found to dominate the excitation for all intensities of interest.

## 2) Multiphoton ionization in a short range potential

Before attacking the difficult problem of a real atom subject to a very strong field one would like to develop an intuition about the new effects caused by such a field by studying model potentials. Even the problem of an electron bound by a model potential and subject to a strong em field has not been solved analytically for any potential. The numerical solution is not an easy task either, the main reason being that strong fields cause ionization and hence any calculation would have to include the continuum spectrum.

Recent experiments on the ionization of atoms have analyzed the outgoing electron energy<sup>5,7</sup>. Several features such as ponderomotive corrections to the ionization potential and above - threshold - ionization -

where the electron absorbs more than the minimum number of photons necessary for ionization - were observed. These features were already qualitatively predicted in earlier work<sup>6,8,9</sup>. In the present work, we will comment on some critical aspects of the strong field, many photon limit. In particular, we solve in this limit a model which represents the atomic potential as a  $\delta$ -function. The analytical results obtained are asymptotically correct as the number of photons goes to infinity. The problem was also solved numerically and very good agreement with the analytical results was obtained even for a few photons.

We approach the ionization problem first in a qualitative fashion. This will allow us to draw a number of conclusions without the necessity of going into complicated equations.

In viewgraph 11, we have plotted a typical binding potential  $V(x)$  and the potential due to an external, linearly polarized, monochromatic field,  $V_{\text{ext}} = Fx \cos(\omega t)$  (atomic units are adopted all through), for each half a period of the field. It is clear that the barrier formed by these two potentials will be up and down in intervals of half a period. When the barrier is down, the electron in the bound state with energy  $\epsilon_0$  must traverse a barrier of width, roughly,  $1 - \epsilon_0/F$ . The time  $\tau_{\text{tunn}}$  to tunnel through that barrier at (imaginary) velocity  $v \sim \sqrt{2\epsilon_0}$  is  $\tau_{\text{tunn}} \sim \sqrt{2\epsilon_0}/F$ . If  $\tau_{\text{tunn}}$  is much smaller than a period of the field, the electron has time to tunnel before the barrier goes up again and the problem is basically static. This is called the adiabatic regime. If, on the other hand,  $\tau_{\text{tunn}}$  is larger than the period of the field, the electron cannot get out of the well before the barrier goes up and we should expect a frequency dependence of the ionization rate. This case is called the multiphoton case. A natural dimensionless parameter is then Keldysh's parameter (viewgraph 12) which is defined as the ratio of the tunneling time to the period of the field,  $\gamma = \tau_{\text{tunn}}/\tau = \sqrt{2\epsilon_0}\omega/F$ . The case  $\gamma \gg 1$  is the multiphoton case and  $\gamma \ll 1$  corresponds to the adiabatic case. Besides  $\gamma$ , there are basically two more dimensionless parameters (viewgraph 18) that characterize the ionization from a general binding potential  $V(x)$ : the ratio between the frequency of the applied field and a typical atomic frequency,  $\omega/\omega_{\text{atom}}$ , whose inverse is the number of photons involved in the process, and the ratio of applied electric field amplitude to a typical atomic field strength,  $F/F_{\text{atom}}$ .

If we consider the relative magnitude of the applied field to the atomic field, we see also from viewgraph 11 that we can distinguish three regions. The first one, which we might call "inside the atom", has  $E_{\text{atom}} > E_{\text{applied}}$ . If this relationship between field strengths did not hold inside the atom, the electron would be ripped off the atom in a few atomic periods. On the other hand, since in a real laser pulse there is always a finite rise time, with such a strong amplitude the atom will ionize before it reaches the peak. The second region is an intermediate one where  $E_{\text{applied}} \sim E_{\text{atom}}$  and finally we have the "outside of the atom" where  $E_{\text{applied}} > E_{\text{atom}}$ . These three regions exist for any typical binding potential subject to a linearly polarized field and consequently any theory that uses a perturbation approach in  $E_{\text{applied}}/E_{\text{atom}}$  or in  $E_{\text{atom}}/E_{\text{applied}}$  cannot be justified because neither assumption holds throughout all space.

In the light of the previous discussion, it is worth pointing out here also the differences between the ionization problem and the scattering problem of an electron in the presence of a strong field. In the scattering problem, the electron can be considered always outside of the atom (external problem) and, under certain conditions, one can assume  $E_{\text{applied}} > E_{\text{atom}}$  uniformly throughout all space. However, the ionization problem presents the extra difficulty that no matter how strong the external field is, there is a region where the electron is initially "inside the atom", where the binding field is larger and where the strong external field approximation breaks down. This difference makes the ionization problem much harder to solve than the scattering problem.

We will now turn to the language of Green's functions, or evolution operators, to put the intuitive description of the previous section into a mathematical form. This allows us to describe our approach and will also enable us later to relate it in a simple way to other works in the literature.

Schrödinger's differential equation for the wave function of an electron subject to a binding potential  $V(x)$  and an external field  $V_{\text{ext}}$  is

$$i \frac{\partial |\psi\rangle}{\partial t} = (T + V + V_{\text{ext}}) |\psi\rangle \equiv H |\psi\rangle$$

where  $T$  is the kinetic energy operator. Similarly, the equation for the Green's function  $G(t, t')$  of our problem is

$$\left( i \frac{\partial}{\partial t} - H \right) G(t, t') = i\delta(t - t') . \quad (2-1)$$

We can also write, formally, the equations for the Green's functions corresponding to the Hamiltonians  $H_0=T+V$  for an electron in the binding potential  $V$ , and to  $H_{\text{ext}}=T+V_{\text{ext}}$  for an electron subject to  $V_{\text{ext}}$  only :

$$\left( i \frac{\partial}{\partial t} - H_0 \right) G_0(t, t') = i\delta(t - t') , \quad (2-1)$$

$$\left( i \frac{\partial}{\partial t} - H_{\text{ext}} \right) G_{\text{ext}}(t, t') = i\delta(t - t') . \quad (2-1)$$

There is clearly a relationship between  $G$  and  $G_{\text{ext}}$  and between  $G$  and  $G_0$ . These relationships are given by the following expressions :

$$G(t, t') = G_0(t, t') - i \int_{t'}^t dt'' G_0(t, t'') V_{\text{ext}}(t'') G(t'', t') \quad (2-2)$$

and

$$G(t, t') = G_{\text{ext}}(t, t') - i \int_{t'}^t dt'' G_{\text{ext}}(t, t'') V(t'') G(t'', t'). \quad (2-3)$$

These integral equations for  $G(t, t')$  are equivalent to Schrödinger's differential equation (1) because they satisfy this equation identically.  $G(t, t')$  is the evolution operator of the system. That means that if  $|\psi(t_0)\rangle$  is the wave function representing the system at time  $t_0$ , the wave function at time  $t$  is given by

$$|\psi(t)\rangle = G(t, t_0) |\psi(t_0)\rangle . \quad (2-4)$$

In particular, one is interested in the transition amplitude for going from an initial state  $|\psi_i(t_0)\rangle$  at time  $t_0$  to a final state  $|\psi_f(t)\rangle$  at time  $t$ . This transition amplitude can be calculated in terms of  $G$  by

$$T_{fi} = \langle \psi_f(t) | G(t, t_0) | \psi_i(t_0) \rangle . \quad (2-5)$$



Unfortunately, the problem of calculating  $G(t, t')$  for an electron subject simultaneously to a binding potential  $V$  and the potential  $V_{\text{ext}}$  due to a plane monochromatic wave, has not been solved analytically for any  $V$ . For calculating the transition amplitudes, one has then to rely on numerical calculations or to some kind of approximation scheme.

The usual way of proceeding is to assume that  $V_{\text{ext}} \ll V$  uniformly in space and calculate  $G$  from an iterative procedure starting from Eq. (2-2). That is, one develops a perturbation series in the applied field strength.

The values of field strength that are now being used in the laboratory preclude this kind of approach. On the other hand, it is not possible to assume that  $V_{\text{ext}} \ll V$  uniformly throughout all space either, because inside the atom, as we saw qualitatively in the previous section, the atomic field is stronger than the applied field.

We can conclude then, that any iterative procedure to calculate  $G$  from either Eq. (2-2) or Eq. (2-3) is of doubtful validity because of the nonuniformity of the relationship between the magnitudes of  $V$  and  $V_{\text{ext}}$  throughout all space.

An alternative way of proceeding is to use either Eq. (2-2) or Eq. (2-3) as a starting point, but with the assumption that  $\omega_{\text{atom}}/\omega \rightarrow \infty$ ; that is, we assume that a large number of photons is involved in the transition.

It proves to be more convenient to use Eq. (2-3). The reason is that the expression for  $G_{\text{ext}}$  is simpler, in general than  $G_0$ . A typical binding potential  $V$  has both a bound and a discrete spectrum making the expression for  $G_0$  very involved. Instead, the Green's function,  $G_{\text{ext}}(t, t')$ , for an electron in the potential (we choose to work in the  $Fx$  gauge)

$$V_{\text{ext}} = Fx \cos(\omega t) \quad (2-6)$$

where  $F$  is the amplitude of the applied field, is well known. Furthermore, because of a result due to Feynman, we can write  $G_{\text{ext}}(t, t')$ , in the  $x$  representation  $G(x, t; x', t') \equiv \langle x | G(t, t') | x' \rangle$ , simply as

$$G_{\text{ext}}(x, t; x', t') = \frac{\exp[iS_{\text{cl}}(x, t; x', t')]}{\sqrt{2\pi i(t - t')}} \quad (2.7)$$

Here  $S_{cl}(x, t; x', t')$  is the classical action of an electron, subject to the potential (2-6), which starts at  $x', t'$  and ends at  $x, t$ . It is given by

$$S_{cl}(x, t; x', t') = \frac{1}{2(t-t')} \{(x-x') - [x_0(t) - x_0(t')]\}^2 - \frac{1}{2} \int_{t'}^t d\tau \dot{x}_0^2(\tau) + x\dot{x}_0(t) - x'\dot{x}_0(t'),$$

expressed in terms of the classical position  $x_0(t)$  of an electron to the external field only; that is,  $x_0(t)$  satisfies  $\ddot{x}_0 = -F(t)$  ( $q = -1$  a.u.).

It proves convenient to work with the following integral equation, derived from Eqs. (2-3) and (2-4) in the  $x$  representation, making use of Eq. (2-7),

$$\psi(x, t) = \psi_0(x, t) - \frac{i^{1/2}}{\sqrt{2\pi}} \int_0^t dt' \frac{1}{\sqrt{t-t'}} \int_{-\infty}^{\infty} dx' \exp[iS_{cl}(x, t; x', t')] V(x') \psi(x', t'). \quad (2.8)$$

The inhomogeneous term in this integral equation for  $\psi(x, t)$  is the wave function  $\psi_0(x, t) \equiv \langle x | \psi_0(t) \rangle$ , where  $\psi_0(t) \rangle$ , which coincides with the bound state at  $t=0$  and which at later times evolves with the Hamiltonian of the electron in the external field only.

We now specialize to the case of electron bound by the potential

$$V(x) = -B\delta(x). \quad (2-9)$$

This potential has only one bound state, of energy  $-B^2/2$  (the ionization potential is then  $I_p = B^2/2$ ), in which the electron is going to be at  $t=0$ . If we specialize Eq. (2-8) for this potential, the  $x'$  integral is straightforward. After setting  $x=0$  in the same equation, we obtain an integral equation, now in time only, for  $\chi(\tau) \equiv \psi(0, \tau)$  :

$$\chi(t) = \chi_0(t) + \frac{i^{1/2}B}{\sqrt{2\pi}} \int_0^t dT \frac{1}{\sqrt{T}} \exp[iS(t, t-T)] \chi(t-T). \quad (2.10)$$

We have made the change of time variables  $T = t - t'$  and we have set  $S(t, t - T) \equiv S_{cl}(0, t; 0, t - T)$ , i.e.,

$$S(t, t - T) = \frac{1}{2T} [x_0(t) - x_0(t - T)]^2 - \frac{1}{2} \int_{t-T}^t d\tau \dot{x}_0^2(\tau). \quad (2-11)$$

Note that we have eliminated in this way the spatial dependence from the problem. From the solution  $\chi(t)$  of Eq. (2-10), we can reconstruct  $\psi(x, t)$  by means of Eq. (2-8). However, as we will see in the next two sections, only  $\chi$  is necessary for computing the ionization rate.

Consider now the ionization of an electron from the  $\delta$ -function potential in a constant (dc) field. The method we use to analyze this problem is somewhat different from the conventional approach and we present it here because it introduces some of the techniques we adopt to analyze the time-dependent field case.

In a constant electric field  $F$ , the classical position of an electron is ( $q=-1$  a.u.)  $x_0 = -Ft^2/2$ . In the potential,  $V = -B\delta(x)$ , the wave function at the site of the potential,  $\chi(t) = \psi(0, t)$  evolves according to Eq. (2-10) with  $S(t, t-T)$  given by Eq. (2-11). Then, simply,  $S(t, t-T) = -F^2T^3/24$  and

$$\chi(t) = \chi_0(t) + \frac{i^{1/2}B}{\sqrt{2\pi}} \int_0^t dT \frac{1}{\sqrt{T}} \exp\left(-\frac{iF^2}{24}T^3\right) \chi(t - T). \quad (2.12)$$

Equation (2-12) is in convolution form. This simply reflects the fact that we are dealing with a time-independent Hamiltonian, the problem is invariant under translations in time and that means that Schrödinger's differential equation can be reduced to an eigenvalue equation and its corresponding integral equation is in convolution form. We can formally solve Eq. (2-12) by using Laplace transforms. Let us define

$$\tilde{\chi}(p) \equiv \int_0^\infty dt \exp(-pT) \chi(t), \quad (2-13)$$

with a similar definition holds for  $\tilde{\chi}_0(p)$ . Then Eq. (2-12) becomes

$$\tilde{\chi}(p) = \frac{\tilde{\chi}_0(p)}{1 - C(p)},$$

with  $C(p)$  given by

$$C(p) = \frac{i^{1/2}B}{\sqrt{2\pi}} \int_0^\infty dt \frac{1}{\sqrt{t}} \exp \left( -pt - \frac{iF^2}{24} t^3 \right). \quad (2-14)$$

The inverse transform is defined by

$$\chi(t) \equiv \frac{1}{2\pi} \int_{-i\infty+\sigma}^{i\infty+\sigma} dp \exp(pt) \tilde{\chi}(p), \quad (2-15)$$

where  $\sigma > \sigma_0$ . The Laplace transform is defined only for  $\text{Re}(p) > \sigma_0$  where  $\sigma_0$  is chosen so that the integral in Eq. (2-13) exists.  $\tilde{\chi}(p)$  is analytically continued from  $\text{Re}(p) > \sigma_0$  to  $\text{Re}(p) < \sigma_0$ . The inverse transform may then be evaluated by closing the  $p$  contour in the left half of the  $p$  plane.  $\chi(t)$  therefore has contributions from the poles (and branch cuts) of  $\tilde{\chi}(p)$ . Specifically, the contribution to  $\chi(t)$  from a pole of  $\tilde{\chi}(p)$  at  $p_0$  is

$$\chi(t) \equiv A \exp(p_0 t) + \dots$$

Note that, by definition all poles of  $\tilde{\chi}(p)$ ,  $p_j$ , have a real part less than  $\sigma_0$ . We may therefore find the ionization rate by finding the positions of the poles of  $\tilde{\chi}(p)$ . (Clearly, the ionization rate  $\omega$  equals the negative real part of  $2p_j$ .) Contributions from  $\chi(t)$  from any branch cuts in the  $p$  plane are ignored for two reasons; first they produce a decay of  $\chi$  like  $t^\beta$ , where  $\beta \geq 1/2$ , so that for  $\omega \ll 1$  and  $t \approx 0(\omega^{-1})$  the exponentially decaying terms dominate  $\chi$ , and, second because these algebraic terms represent contributions to  $\chi$  from free wave packets that are spreading and propagating away from  $x=0$  and not contributions from a "quasibound" wave function. With "smooth" initial conditions,  $\tilde{\chi}_0(p)$  has no poles so that we may write

$$1 - C(p_j) = 0 \quad (2.16)$$

Our treatment is analogous to Landau's<sup>11</sup> treatment of the decay of plasma oscillations where Eq. (2-16) takes the role of the "dispersion

relation". By assuming that the initial conditions are smooth, we obtain a decay rate that does not depend on them. If we take the initial condition to be the bound state of the  $\delta$ -function model,  $\tilde{\chi}_0(p)$  has no poles.

Equation (2-16) is a transcendental equation for the  $p_j$ 's and, unfortunately, the integral  $C(p)$  is not available in a closed form. We are mainly interested in the limit when  $F \ll B^3$  (or in physical terms the applied electric field is smaller than the atomic field when the electron is in the bound state) in this limit  $C(p)$  can be evaluated asymptotically near  $p_j$  via the method of steepest descents. When  $F=0$ , there is only one pole  $p_0=iB^2/2$  and consider the integral for  $C(p)$ , Eq. (2-14), in the complex  $t$  plane. The steepest-descent path for the exponential argument  $(-i\epsilon t - iF^2 t^3/24 - 1/2 \ln t)$  from  $t=0$  is (for  $\epsilon > 0$ ) along the negative imaginary axis to the saddle point at  $t_0 = -i[8(\epsilon + i\omega/2)/F^2]^{1/2}$  and then in the direction of the increasing  $\text{Re } t$ . This path is illustrated in viewgraph 19 where the scaled variable  $z \equiv tF/\epsilon_0^{1/2}$  has been introduced in order to generate a universal curve. The path of integration consists of two parts, 0 to  $t_0$ , the dominant part  $C_D$ , and  $t_0$  to  $\infty$ , the subdominant exponentially small part,  $C_S$ . If we set  $t = -is$  along the first path,

$$\begin{aligned} C(p) &= \frac{B}{\sqrt{2\pi}} \int_0^{s_0} ds \frac{1}{\sqrt{s}} \exp \left( - \left( \epsilon + i\frac{\omega}{2} \right) s + \frac{F^2 s^3}{24} \right) \\ &+ \frac{i^{1/2} B}{2\pi} \int_{t_0}^{\infty} dt \frac{1}{\sqrt{t}} \exp \left( - i\epsilon t + \frac{\omega}{2} t - \frac{iF^2}{24} t^3 \right) \\ &= C_D \left( \epsilon + i\frac{\omega}{2} \right) + C_S \left( \epsilon + i\frac{\omega}{2} \right), \end{aligned}$$

where  $s_0 = it_0 \equiv \sqrt{8\epsilon_0}/F$ . We note that ( $\omega \ll \epsilon$  and  $\epsilon \equiv \epsilon_0$ )

$$\begin{aligned} C_S(\epsilon_0) &\equiv 0 \left( \exp \left( - i\epsilon_0 t_0 - \frac{iF^2}{24} t_0^3 \right) \right) \\ &\equiv 0 \left( \exp \left( - \frac{2}{3} \frac{B^3}{F} \right) \right) \ll 1, \end{aligned}$$

where we have used  $\epsilon_0 = B^2/2$ . We therefore look for solutions of the equation

$$1 - C_D \left( \epsilon + i \frac{\omega}{2} \right) = 0 \quad (2-17)$$

Equation (2-17) can be solved by expansion of the term  $\exp(-F^2 s^3/24)$  in the integrand to obtain an asymptotic series for  $\epsilon$  in  $F/B^3$ . From Eq. (2-16), we obtain for  $\epsilon$  and  $\omega$

$$\epsilon = \frac{B^2}{2} \left( 1 + \frac{5}{4} \frac{F^2}{B^6} + \dots \right) \quad (2-18)$$

$$\omega = 0$$

One may easily verify that  $\omega$  is identically zero in Eq. (2-17). Therefore  $\omega$  is determined by including  $C_S$ ; specifically

$$\frac{\omega}{2} \frac{\partial C_D}{\partial \epsilon} = - \text{Im} C_S(\epsilon) \quad (2-19)$$

$C_S(\epsilon)$  can be determined by noting that the dominant contribution to the integral comes from the integration over half the saddle point at  $t_0$ . Evaluating  $C_S$  and  $\partial C_D/\partial \epsilon$  to lowest order we obtain the ionization rate

$$\omega = B^2 \exp \left( - \frac{2B^3}{3F} \right). \quad (2-20)$$

In this evaluation it is sufficient to use  $\epsilon \approx B^2/2$  in Eq. (2-20).

The separation of the integral into  $C_D$  and  $C_S$  is a crucial part of our analysis. The real frequency  $\epsilon$  (the energy eigenvalue plus its shift) comes from  $C_D$  and it is given by an asymptotic series in  $F/B^3$ . The imaginary frequency  $\omega/2$  (half of the ionization rate) comes solely from including  $C_S$  and is exponentially small in the parameter  $F/B^3$ . Since the exponentially small terms are in the asymptotic sense "beyond all orders" in  $F/B^3$  it is essential that the solution of Eq. (2-16) (including only  $C_D$ ) gives a  $\omega$  that is identically zero. The separation of  $C$  into  $C_D$  and  $C_S$  enables us to separate the contributions to  $\omega$  from the corrections to  $\epsilon$  which, although larger in the asymptotic sense, are irrelevant. A similar separation of the problem occurs in the many-photon limit treated in the next section.

We will study the problem of the ionization of an electron bound by the same single  $\delta$ -function potential treated in the last section, but now subject to an external, monochromatic electric field, as represented by the potential, Eq. (2-6). The integral equation for  $\chi$ , the wave function at the site of the potential  $V(x)=-B\delta(x)$ , is again given by Eq. (2-10). Now, however, due to the time dependence of  $V_{\text{ext}}$ , this equation is no longer in convolution form, a reflection of the fact that the corresponding Schrödinger differential equation cannot be reduced to an eigenvalue equation.

We look for a solution of the integral equation (2-10) in the eikonal form

$$\chi(t) = \exp \left( i \int_0^t d\tau \epsilon(\tau) \right), \quad (2-21)$$

with  $\epsilon(\tau)$  complex. Intuitively, one can think of Eq. (2-21) as a "generalized" of the inverse Laplace transform, Eq. (2-15), useful for a slowly time-varying Hamiltonian. We expect  $\epsilon$  to vary correspondingly slowly in time. Note that the exponent  $\int d\eta \epsilon(\eta)/\omega$  is proportional to the number of photons,  $\epsilon/\omega$ , and is indeed proportional to a large quantity that is slowly varying, in analogy with the situation in WKB or in geometrical optics.

Substituting the assumed form, Eq. (2-21) into the governing integral Eq. (2-10), and neglecting the inhomogeneous term, as for the time-dependent case, we obtain

$$1 = \frac{i^{1/2}B}{\sqrt{2\pi}} \int_0^t dT \frac{1}{\sqrt{T}} \times \exp \left( i \left( S(t, t-T) - \int_{t-T}^t d\tau \epsilon(\tau) \right) \right), \quad (2-22)$$

from which we wish to develop an asymptotic expression for  $\epsilon(t)$ . In order to make progress, we assume that, for sufficiently weak fields  $F$ ,  $\epsilon$  is nearly real. Then the argument of the exponential is large and nearly imaginary. This is the case since the two terms in the argument

$$\int_{t-T}^t \epsilon(\tau) d\tau \cong B^2/\omega \quad \text{and} \quad S \cong F^2/\omega^3$$

are both assumed large. The ratio of these two terms is proportional to  $\gamma^2$ , where  $\gamma \equiv B\omega/F$  is Keldysh's adiabatic parameter, defined in above. Because of the  $T^{-1/2}$  factor in the integrand, the largest contribution to the integral comes from  $T=0$ . This forms the basis for approximation as follows: We can write, for small  $T$ ,

$$\int_{t-T}^t \epsilon(\tau) d\tau \cong \epsilon(t)T \quad (2-23)$$

We note, further, that both  $S$  and its derivative with respect to  $T$  approach zero as  $T$  approaches 0 (cf Eq. (2-11)). These observations enable an evaluation of the dominant contribution to the integral in Eq. (2-22). The resulting evaluation of the "dispersion relation" yields, to lowest order,

$$\epsilon(t) = B^2/2. \quad (2-24)$$

In order to improve the result and calculate the ionization rate, the rest of the time integration needs to be included. Proceeding in the same spirit as in the time-dependent case, we write

$$\epsilon(t) = \epsilon_R(t) + i\delta(t), \quad (2-25)$$

where the result, Eq. (2-24), is the first approximation to the real frequency  $\epsilon_R(t)$ . The method for improving the result is analogous to the dc procedure. The contribution to the integral for small  $T$  is evaluated in terms of the unknown, exact  $\epsilon(t)$ , whereas the remainder of the integration, to be evaluated via steepest descents, is computed using only the lowest-order result, Eq. (2-24), for the eigenvalue.

The difficulty of the problem is enhanced over the dc problem because the integral equation is no longer in convolution form and there are many saddle-points because of the periodicity of the field. The detailed treatment of the problem is given in Ref. 11. However, we are interested in calculating the time average ionization



$$w = \lim_{T \rightarrow \infty} \int_0^T dt \delta(t) \quad (2-26)$$

It may be shown that contribution from  $w$  comes from the saddle-point integrations and further one need only keep the lowest order result for  $\epsilon$ , Eq. (2-24), in evaluating these integrals.

The general expression for  $w$  is complicated for arbitrary  $\gamma$ , but has the general feature that the ionization rate is a sum of processes involving in integral number  $\sigma$  of photons, where  $\sigma \geq B^2/2\omega^2 + F^2/4\omega^3$ . This is the so-called above threshold ionization. The ionization potential as well as the ponderomotive potential  $F^2/4\omega^2$  must be overcome by the ejected electron. See viewgraph 22.

In the limit  $\gamma \rightarrow 0$  we obtain

$$w = (3F/\pi B)^{1/2} B^2 \exp\left(-\frac{2}{3} \frac{B^3}{F}\right) \quad (2-27)$$

It is instructive to obtain this result in another way. If in the dc result Eq. (2-20) we let  $F$  have the time dependence  $F \rightarrow F \cos(\omega t)$  and treat  $\omega$  small, the system tracks the field adiabatically and we can average the expression for  $w$  over one period of the applied field to obtain  $w_{\text{avg}}$ . (See viewgraph 22).

If  $F \ll B^3$  the integral may be evaluated by the saddle-point method where we obtain two Gaussian contributions in the period  $2\pi/\omega$ . This corresponds physically to the fact that we have an ionization "burst" each time the barrier is low. Calculating  $w_{\text{avg}}$  in this way recovers the previous result.

For the case  $\gamma \rightarrow 0$  we find

$$w \equiv \frac{2}{\pi} \frac{\omega B}{k_s} \left( \frac{F e^{1/2}}{2\omega B} \right)^{2\sigma} \quad (2-28)$$

$\sigma \approx B^2/2\omega$  is the minimum number of photons to reach the continuum and  $k_e^2/2 = -(B^2/2)(1 + 1/2\gamma^2) + 1/2\gamma^2 + s\omega$  is the ejected electron energy.

In addition to the analytic work, a numerical solution of the integral equation was undertaken. Viewgraph 23 shows a plot of  $|\chi(t)|^2$  for the parameters there indicated. The slope of  $|\chi|^2$  versus  $t$  is directly proportional to the bound state probability.

Viewgraph 24 and 25 show log-log plots of the ionization rate versus the value of  $F$  for the time dependent field normalized to the atomic field strength,  $F_{\text{atm}} = B^3$ . The curve is an evaluation from the asymptotic formula while the dots are results obtained from numerical solution of the integral equation. In viewgraph 24,  $\omega = .5$  and  $B = 1.59$ . The minimum of photons required for ionization as  $F \rightarrow 0$  is 3, while for the larger fields strengths an additional photon is needed to overcome the increasing ponderomotive potential,  $F^2/4\omega^2$ . The comparison is quite good even though few photons are involved. In viewgraph 25  $\omega$  has been reduced to 0.25. The minimum number of photons is now seven and increases to fifteen at the highest fields. The additional structure is due to the change of  $\sigma$  to  $\sigma + 1$  photons with relatively little change the field.

### 3) Charge states of many electron atom in strong fields

As we have discussed, modern laser systems can deliver electric field strengths near the focal region on the order of or larger than the nuclear electric field as seen by the outer shell electrons. On the other hand, such intensities are available only at photon energies much smaller than typical binding energies, so that ionization requires absorption of a large number of photons. Recent experiments on noble gases<sup>12-15</sup> investigating multiphoton ionization (MPI) show markedly different behavior depending on whether an intermediate state is excited (resonant ionization) or not (nonresonant), with the intensity required for observation of a given charge state (the threshold intensity) orders of magnitude lower in the former case. In contrast, a feature of the nonresonant experiments is the apparent absence of shell effects. Indeed, in refs ? and ? the authors note (see viewgraph 26) that the observed threshold intensities depend very little on the details of the structure of

the particular atom under study, but depend only on the binding energy of the extracted electron.

We limit ourselves here to a discussion of nonresonant ionization phenomena. Because of the high applied fields used in recent experiments, any theory purporting to explain the data has to be non-perturbative in character. In reference 12, the interpretation was based on the Keldysh single-electron model which describes MPI from the ground state of hydrogen ignoring effects of intermediate states. The multi-electron nature of the problem could, in principle, be addressed numerically at the level of a Hartree-Fock calculation. However, for a system such as a Xenon atom in the presence of a strong field this appears to be prohibitive in practice. A simpler approach, motivated by the apparent absence of shell effects in the experimental results and by the large number of electrons is to use a statistical model, such as the Thomas-Fermi model (see viewgraphs 27 and 28).

In the Thomas-Fermi model, the electronic cloud is considered to be a zero temperature Fermi gas whose density is calculated by filling the available phase space volume, taking account of the Pauli exclusion principle (viewgraphs 30, 31 and 32). The density is used in Poisson's equation to compute the self-consistent electrostatic potential of the atom. Alternatively, the Thomas-Fermi model can be shown to follow from the Hartree-Fock model upon the neglect of exchange effects and by the semiclassical computation of the single-particle wave functions. For a large  $Z$  atom, exchange and quantum corrections are both of order  $Z^{-2/3}$  compared to the terms retained in the Thomas-Fermi model (viewgraph 29).

One apparent inaccuracy of the model is its prediction of a weak, algebraic decay of electron density with radius as  $r \rightarrow \infty$  for a neutral atom. However, for ions, the case of most interest here, the density is more realistically confined to a finite radius.

The disparity between the laser frequency and a characteristic atomic frequency suggests a straightforward modification of the model to study atoms in the presence of a low frequency electromagnetic field viz. : We add here an electric field  $E(x, t) = \hat{2}F(x, t) \cos \omega t$  to the model. Here  $\omega$

is wave frequency and  $F$  a slowly varying amplitude. The combined potential  $\phi(x, t) = \phi_{\text{atomic}} + Fz$  has an instantaneous saddle point at  $z = z_s$  and a value  $-\phi_s$  there (both  $z_s$  and  $\phi_s$  are to be determined). Those electrons with energy  $-\epsilon > -\phi_s$  are ionized with a single field period near the peak of the field so that it is sensible to freeze  $F$  in our calculations.

Within the model, it is only through increases in intensity, that is,  $F$ , that additional ionization occurs. The contribution to ionization from tunneling by electrons of energy  $-\epsilon < -\phi_s$  through the barrier is ignored in the semiclassical limit. However, we will find that, for the short pulse experiments to which we compare, tunneling is negligible.

An estimate of the tunneling contribution to the ionization can be obtained from the well-known Landau formula. This formula gives the tunneling rate for an electron through a barrier formed by a coulomb potential and a D.C. field,  $w = 8\omega_a(F_a/F) \exp(-2F_a/3F)$  in atomic units. Here,  $F_a$  is the coulomb field exerted on the electron in the ground state and  $\omega_a$  is the orbital frequency. (This formula is valid when the barrier width is large compared to the de Broglie wavelength of the electron.) It is also known that, in the non-coulomb case, only the pre-exponential part of this formula will change while the exponent remains the same. We can then rely on this formula for an order of magnitude estimate of the tunneling rate in our problem.  $\omega_a$  and  $F_a$  for our ions can be estimated as  $I_p$  and  $(2I_p)^{3/2}$ , respectively. Clearly, the most favorable situation for tunneling is in the strongest possible field (largest  $F/F_a$ ). In Augst et al.'s experiment, this occurs at the highest intensities ( $\sim 10^{16}$  W/cm<sup>2</sup>), for which the ionization potential  $I_p \sim 100$  eV. For a picosecond pulse, we have  $\omega\tau \sim 10^{-4}$  for the tunneling contribution, which is negligible.

Since tunneling is negligible, the width of the above-threshold-ionization (ATI) spectrum is not resolved. This width arises, in the adiabatic (tunneling) limit, from the fact that tunneling ionization occurs not only at phases  $\phi_n = n\pi$  corresponding to field maxima, but for small interval  $\Delta\phi$  around  $\phi_n$ . In the current situation, the width is likely set by other uncertainties. Furthermore, because all of the ejected electrons are freed from the atom at a local field maximum, they subsequently gain average oscillatory energy  $\langle mv_{\text{osc}}^2/2 \rangle = m(eF_{\text{ioniz}}/m\omega)^2/4$  where  $F_{\text{ioniz}}$  is the field amplitude at the time of ionization and  $\langle \rangle$  indicates an average

over the field period  $2\pi/\omega$ . Since these ponderomotive effects are simply additive, we ignore them below.

In detail, then, our system consists of  $Q_{el}$  electrons around a nucleus of charge  $Z$ , subject to an external potential  $V_{ext} = -Fz$ , where the  $z$ -axis is chosen along the direction of the field. (Atomic units are used throughout.) If we are a position  $r$  and the maximum available energy of a single electron in the atom is  $-\epsilon$ , then, classically, the momentum of the fastest electron at  $r$  is

$$p_f = \sqrt{2[\phi(r) + Fz - \epsilon]}$$

where  $-\epsilon$  is the energy at the separatrix and  $-\phi$  is the effective potential seen by the electron.

From the well-known semiclassical phase-space quantization argument, the electron number density is

$$n(r) = \frac{p_f^3}{3\pi^2} = \frac{2^{3/2}}{3\pi^2} (\phi + Fz - \epsilon)^{3/2} \quad (3-1)$$

The electron density is taken to be zero outside the region delimited by the separatrix (outside the classical region). Equation (3-1) is two dimensional, due to the azimuthal symmetry around the field axis. This necessitates a numerical investigation.

It is convenient to change the dependent variable  $\phi$  to  $\chi$ , where  $\phi + Fz = (Z/r)\chi$ , and the independent variable  $r$  to the Thomas-Fermi scaled coordinates  $r = bZ^{-1/3}x$  ( $b \equiv 2^{-7/3}(3\pi)^{2/3}$ ). With these changes, Poisson's equation for  $\phi$ , with the electron density Eq. (3-1) written in spherical coordinates ( $\mu \equiv \cos\theta$ ) is

$$\frac{\partial^2 \chi}{\partial x^2} + \frac{1}{x^2} \frac{\partial}{\partial \mu} [(1 - \mu^2) \frac{\partial \chi}{\partial \mu}] = \begin{cases} [\chi - (\epsilon b/Z^{4/3})x]^{3/2}/\sqrt{x}, & \text{if } x < x_s(\mu) \\ 0, & \text{otherwise,} \end{cases} \quad (3-2)$$

where  $x_s(\mu)$  is the separatrix curve (the curve that satisfies  $\chi - \epsilon Z^{-4/3}bx = 0$ ). For  $r \rightarrow 0$  ( $x \rightarrow 0$ ) the nucleus is unscreened, and for  $r \rightarrow \infty$  ( $x \rightarrow \infty$ ) we

determine the boundary condition by asking the total number of electrons to be  $Q_{el}$ . In terms of  $\chi$ , these conditions result in

$$\begin{cases} \chi(0, \mu) = 1 \\ \chi \rightarrow Q/Z + (F/Z^{5/3})b^2x^2\mu \text{ for } x \rightarrow \infty \end{cases}$$

where  $Q \equiv Z - Q_{el}$  is the total charge of the atom (nuclear + electronic). The electronic charge,  $Q_{el}$ , is in turn determined from  $\chi$  by

$$\frac{Q_{el}}{Z} = \frac{1}{2} \int_{-1}^1 d\mu \int_0^{x_s(\mu)} dx x^{1/2} \left( \chi - \frac{\epsilon b}{Z^{4/3}} x \right)^{3/2}. \quad (3-4)$$

We can also obtain an expression for the dipole moment,  $D_z$ , induced by the external field  $F$ . In analogy with Eq. (3-4),

$$D_z = \langle z \rangle = \frac{\int dV n(r) z}{\int n(r) dV} = \frac{Z^{2/3} b}{Q_{el}} \frac{1}{2} \int_{-1}^1 d\mu \mu \int_0^{x_s(\mu)} dx x^{3/2} \left( \chi - \frac{\epsilon b}{Z^{4/3}} x \right)^{3/2}. \quad (3-5)$$

In Eq. (3-2), with boundary conditions given by Eqs (3-3, 3-4),  $\epsilon$  appears only in the combination  $\epsilon Z^{-4/3}$  and the field strength  $F$  appears only in the combination  $FZ^{-5/3}$ . It is clear then that  $\epsilon Z^{-4/3} = g(FZ^{-5/3})$ , where  $g$  is a function which is independent of  $Z$ ; that is, universal for all (heavy) atoms. In the same way, from Eq. (3-4)  $Q_{el}/Z$ , and hence  $Q/Z = 1 - Q_{el}/Z$ , are universal functions of  $\epsilon Z^{-4/3}$ , and hence of  $FZ^{-5/3}$ , i.e.,  $Q/Z = h(FZ^{-5/3})$ . The same reasoning holds for the induced dipole moment  $D_z Z^{1/3}$  given by Eq. (3-5). It is, therefore, sufficient to calculate the charge states, induced dipole moment, and  $\epsilon$  curves versus applied field strength for  $Z = 1$  only, which produces the universal curves for all atoms.

We choose to solve Eq. (3-2) for  $\chi(x, \mu)$  numerically by a relaxation method. We put an extra  $\partial x / \partial t$  in Eq. (3-2) and obtain then a parabolic equation whose solution will converge to the solution of Eq. (3-2) as  $t \rightarrow \infty$ . To solve the two-dimensional partial differential equation, we use an alternate-direction-implicit scheme. We take as initial conditions for  $\chi$ , the well-known Thomas-Fermi function  $\chi_{TF}$  for the bare atom ( $F = 0$ ). (This function satisfies  $\chi_{TF} = \chi_{TF}^{3/2} / x^{1/2}$ ). We take  $\epsilon = 0$  initially. At each subsequent time step we find  $\epsilon$  as follows : because of the azimuthal

symmetry, we know that the saddle point of  $\phi$  (hence  $\chi/x$ ) is at a point  $z = z_s$  along the  $z$ -axis. We find this point numerically by solving  $\partial(\chi/x)/\partial z = 0$ . Having  $z_s$ , we can calculate the energy at the separatrix as  $\epsilon = Z^{4/3}\chi(z_s, 1)/(bz_s)$ .

Once we know  $\epsilon$ , we can generate the rest of the separatrix curve  $x \equiv x_s(\mu)$  by solving  $\chi(x, \mu) - \epsilon Z^{-4/3}bx = 0$  numerically for  $x = x_s(\mu)$ . Knowing  $x_s(\mu)$ , we can construct the updated electron density in the right-hand side of Eq. (3-2) and solve again for  $\chi$  repeating the whole process until we achieve self-consistency.

The resulting universal curves for the total charge of the ion,  $Q/Z$ , and dipole moment,  $D_z Z^{1/3}$ , versus field,  $FZ^{-5/3}$  are presented in viewgraph 30 and 31, respectively.

The physical meaning of  $\epsilon$  is, as mentioned before, the maximum energy of a single bound electron in the presence of the external field. The amount of energy required to extract an electron from an ion  $X^{+q}$  for  $F = 0$  is the ionization potential,  $I_p$ , of that charge state  $X^{+q}$ . It proves to be more convenient for comparison with experimental data to plot the field required to produce a certain charge state versus the ionization potential of that charge state, rather than versus  $\epsilon$ . For that, we need to calculate the ionization potential of the Thomas-Fermi atom or ion of a certain charge and also the field required to produce that charge. The resulting universal curve of scaled intensity,  $IZ^{-10/3}$ , plotted versus scaled ionization potential,  $I_p Z^{-4/3}$  is shown in viewgraph 34.

We compare our results with the data presented in Ref. 13. There the authors summarized the results of their experiments on multiphoton ionization of He, Ne, Ar, Kr, and Xe by high-intensity 1  $\mu$ m, 1 psec laser pulse in a plot of the threshold intensity for the appearance of a charge state versus the ionization potential of the previous charge state. They defined the threshold or appearance intensity as the intensity at which a small number of ions is produced, which for the lowest charge states corresponded to a yield of  $\sim 5 \times 10^{-3}$  (compared with  $< 10^4$  for tunneling). In our semiclassical model, we cannot calculate the field which yields small ionization probability but we can and do calculate the field for an

ionization probability equal to 1. This provides an upper bound for the appearance intensity measured in experiments.

If we replot viewgraph 34 for  $Z = 2, 10, 18, 36$  and  $54$ , and display the results in experimental units a direct comparison can be made with data, as shown in viewgraph 35. The sequence of theoretical curves progresses from left to right for increasing  $Z$ , as do the experimental points. It is clear that the model gives the right scaling with the atomic number  $Z$  and, moreover, that it is in quite good agreement with the observed appearance intensity of a given charge data.

We remark that the interpretation of their data by the authors of Ref. 13 is similar in spirit to our approach. However, they ignore self-consistent deformation of the charge cloud in computing  $\epsilon$  and use the unperturbed values of the energy levels in their computation of the field strength required for ionization. The fact that the charge cloud is indeed only slightly deformed at those field strengths which lead to ionization is a result of our self-consistent calculation, as seen from the maximum value  $D_Z Z^{1/3} \approx .094$  in viewgraph 33.

We note that Perry et al.'s results for threshold using  $1\mu\text{m}$  radiation are as much as an order magnitude lower than those of Augst et al. One relevant difference between the two experiments may be that the Keldysh adiabaticity parameter  $\gamma \equiv \omega(2m\chi)^{1/2}/qE_0$ , with  $\omega$ ,  $E_0$  the laser frequency and amplitude, respectively,  $\chi$  the ionization potential, and  $q$ ,  $m$  the electron charge and mass, respectively. When  $\gamma < 1$ , the ionization rate is expected to be independent of field frequency and to be calculable using a static model. It ranges from 1-2 in the former case and from .17 at the highest intensities to .95 at the lowest intensities for Augst et al.

In conclusion, the intensity required to reach a specified charge state by non-resonance multiphoton ionization of high  $Z$  atoms has been obtained by employing a statistical model of the atom subject to a constant electric field.



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- Outline

- Motivation

- Development of very powerful short-pulse lasers
- Progression of X-ray lasers toward shorter wavelength
- Various particle acceleration and frequency upshifting schemes

- Essential difficulties and our approach

- $\omega \ll \omega_{\text{atom}}$ , but  $F/F_{\text{atom}}$  not very small.
- Perturbation theory is inappropriate. (Very high order calculation of unknown validity.)
- Occupation number is an adiabatic invariant. Isomorphic to above barrier reflection problem.
- Our technique is asymptotic in  $\omega/\omega_{\text{atom}}$  and makes use of deformation into the complex time domain to capture these small effects.

- Two-Level System
  - Simplest prototypical system
  - Elucidates asymptotic techniques applicable to more realistic systems
- Multiphoton Ionization of a particle in a Short Range Potential
  - Ponderomotive Shift in Ionization Potential
  - ATI peaks
  - Comparison of analytic theory and numerical solutions
- Application of the Thomas-Fermi Model to MPI of many electron atoms and Ions
  - Comparison of predictions of TF model with recent experimental results

- The two-level atom

- R. Duvall's thesis

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- Simple hamiltonian

$$i \frac{da}{dt} = \mathbf{H}(t) \cdot \mathbf{a} \quad ,$$

of the explicit form

$$i \frac{d}{dt} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \begin{pmatrix} E_1 & F \sin \omega t \\ F \sin \omega t & E_2 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} \quad .$$

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[Viewgraph 3](#)

- Can be written equivalently as a second order system

$$\frac{d^2 y}{d\tau^2} + Qy = 0,$$

where

$$Q \equiv \left(\frac{\Delta}{\omega}\right)^2 \left[1 + \left(\frac{F}{\Delta}\right)^2 \sin^2 \tau + i \frac{\omega F}{\Delta^2} \cos \tau\right],$$

with

$$\Delta \equiv \frac{(E_2 - E_1)}{2}.$$

- Our solution is asymptotic in

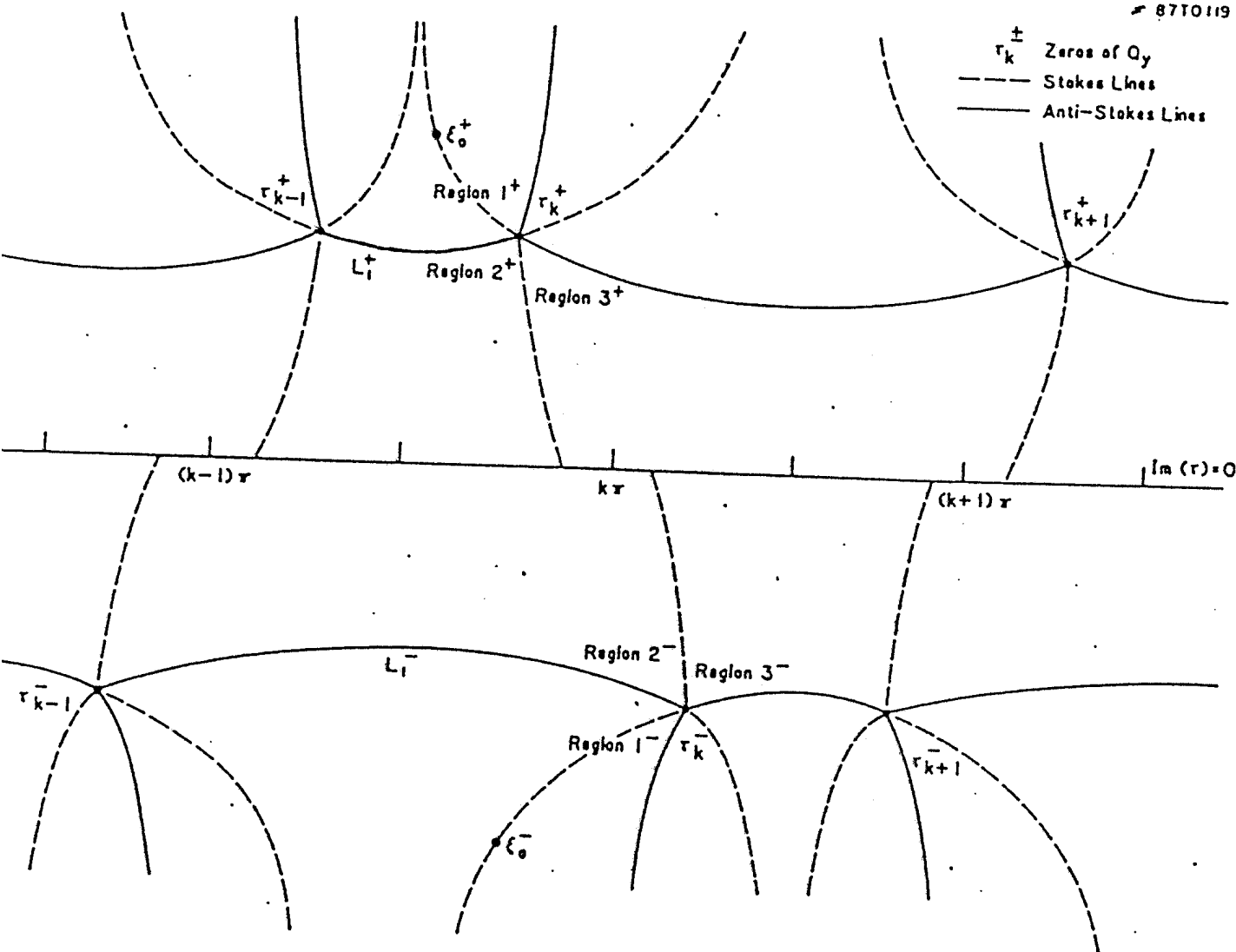
$$\frac{\Delta}{\omega} \gg 1,$$

rather than perturbative in  $F/\Delta$ .

- Because  $\Delta/\omega \gg 1$ ,  $Q \gg 1$  on the real time axis)

Viewgraph 4

87T0119



Viewgraph 5 :

Stokes and anti-Stokes line structure for  $Q_y$  in the complex  $\tau$  plane for an interval surrounding  $\tau = k\pi$ , where  $k$  is taken even. The structure repeats as  $\tau$  changes by  $2\pi$ .

- We represent  $y$  asymptotically in the WKB form

$$y \sim c_k^+(\tau_k^+, \tau) + c_k^-(\tau, \tau_k^-) \quad , \quad |\tau - k\pi| < \pi .$$

where

$$(a, b) \equiv Q^{-1/4}(\tau) \exp(+i \int_a^b Q^{1/2} d\tau) \quad ,$$

- We compute the change in  $c_k$  over one period as we cross the Stokes' lines.
- Denoting  $\mathbf{c}_k = \begin{pmatrix} c_k^+ \\ c_k^- \end{pmatrix}$ , we eventually obtain a matrix relating  $\mathbf{c}_{k+2}$  to  $\mathbf{c}_k$ ,

$$\mathbf{c}_{k+2} = \mathbf{M} \cdot \mathbf{c}_k \quad .$$

where

$$\mathbf{M} = \begin{pmatrix} \exp(i2\theta_0) & 2\varepsilon \sin \theta_0 \exp(i\theta_0) \\ 2\varepsilon \sin \theta_0 \exp(-i\theta_0) & \exp(-i2\theta_0) \end{pmatrix} \quad .$$

is expressed in terms of

$$\varepsilon \equiv (\tau_k^-, \tau_k^+) ,$$

[Viewgraph 6](#)

and of the (evidently real) phase advance per half period of the applied field

$$\theta_0 \equiv (0, \pi) = \frac{\Delta}{\omega} \int_0^\pi \left[ 1 + \left( \frac{F}{\Delta} \right)^2 \sin^2 \tau + i \frac{\omega F}{\Delta^2} \cos \tau \right]^{\frac{1}{2}} d\tau \quad ,$$

- $\varepsilon$  has the asymptotic forms

$$\begin{aligned} |\varepsilon| &\sim \left( \frac{e F}{4 \Delta} \right)^{2 \frac{\Delta}{\omega}} \quad , & F/\Delta &\ll 1 \quad , \\ &\sim \exp\left(-\frac{\pi \Delta \Delta}{2 \omega F}\right) \quad , & F/\Delta &\gg 1 \quad , \end{aligned}$$

- $\theta_0$  has the asymptotic forms

$$\begin{aligned} \theta_0 &\sim \pi \frac{\Delta}{\omega} \left[ 1 + \frac{1}{4} \left( \frac{F}{\Delta} \right)^2 \right] \quad , & F/\Delta &\ll 1 \quad , \\ &\sim 2 \frac{F}{\omega} \quad , & F/\Delta &\gg 1 \quad . \end{aligned}$$

- The matrix which evolves  $\mathbf{a}_k$  over an integral number  $N$  of periods is, finally,

$$\mathbf{a}_{k+2N} = \mathbf{M}^N \cdot \mathbf{a}_k \quad .$$

[Viewgraph 7](#)



- Assuming  $a^-(0) = 0$ ,  $a^+(0) = 1$ , setting  $P \equiv |a_{k+2N}^-|^2$  and making the replacement  $N \rightarrow \omega t/2\pi$ , we obtain the result, for  $\omega t \sim \varepsilon^{-1}$ ,

$$P \simeq \frac{u^2}{(\omega_{\text{res}} - (2p+1)\omega)^2 + u^2} \sin^2 \left\{ \frac{1}{2} [(\omega_{\text{res}} - (2p+1)\omega)^2 + u^2]^{\frac{1}{2}} t \right\}$$

where we have defined  $u \equiv (2\omega/\pi)|\varepsilon|$  and  $\omega_{\text{res}} \equiv (2\omega/\pi)\theta_0$  where  $l = 2p + 1$ , and  $p$  is an integer.

- The resonance condition is

$$\omega_{\text{res}} - (2p+1)\omega = 0 \quad .$$

- In the small field limit ( $F/\Delta$  small), we have

$$u \sim \frac{2\omega}{\pi} \left( \frac{eF}{4\Delta} \right)^{2\frac{\Delta}{\omega}} ,$$

$$\omega_{\text{res}} \sim 2\Delta \left( 1 + \frac{1}{4} \left( \frac{F}{\Delta} \right)^2 \right) \quad .$$

(These results are equivalent to those obtained perturbatively by Shirley.)

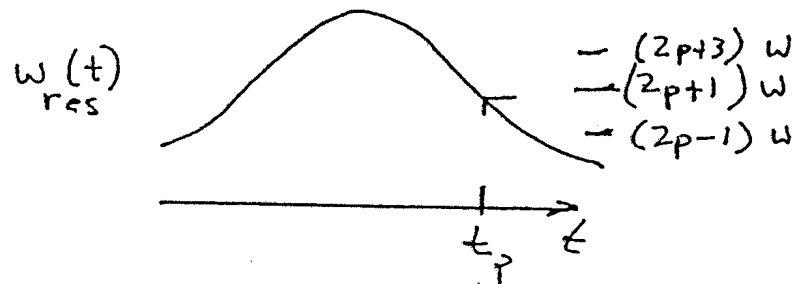
- For the case of large fields ( $F/\Delta$  large) we have

$$u \sim \frac{2\omega}{\pi} \exp\left[-\frac{\pi \Delta \Delta}{2 \omega F}\right] \quad ,$$

$$\omega_{\text{res}} \sim \frac{4F}{\pi} \quad .$$

Viewgraph 9

- An Effect of Finite Pulse Length on high order resonances



- Because of the intensity dependence of  $\omega_{res}$  the condition

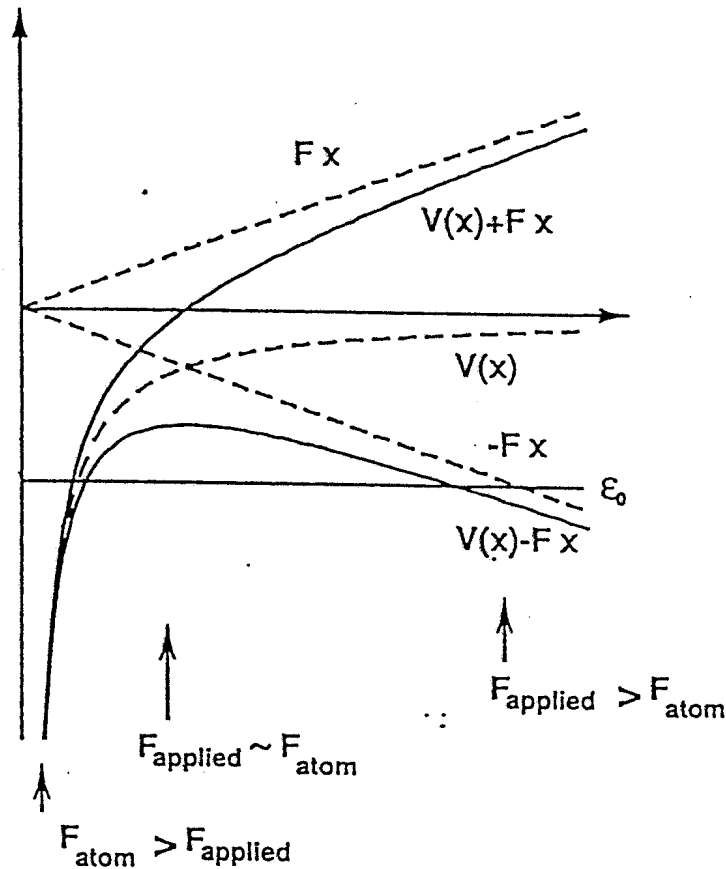
$$\omega_{res} - (2p + 1)\omega = 0,$$

for resonance of a given order  $p$  is satisfied precisely at only a single time  $t_p$ .

- If  $p \gg 1$ , then it can be that many resonances are passed through as the applied pulse evolves.
- In the limit in which successive resonances contribute with random phases one obtains an average rate of excitation

$$R = \frac{\pi u^2}{4\omega}.$$

- There are several parameters characterizing MPI



- The electron in the bound state with energy  $\epsilon_0$  “sees” a barrier of width, roughly,  $l \sim \epsilon_0/F$ .
- The time  $\tau_{\text{tunn}}$  to tunnel through that barrier at (imaginary) velocity  $v \sim \sqrt{2\epsilon_0}$  is  $\tau_{\text{tunn}} \sim \sqrt{2\epsilon_0}/F$ .

- The ratio of the tunneling time to the period of the field

$$\gamma \equiv \tau_{\text{tunn}}/\tau = \sqrt{2\epsilon_0}\omega/F.$$

is called the **Keldysh parameter**.

- from the original work of L.V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys. JETP 20, 1307 (1965)].
- cf, also A.M. Perelomov, V.S. Popov and M.V. Terentev, Zh. Eksp. Teor. Fiz. 50, 1393 (1966) [Sov. Phys. JETP 23, 924 (1966)].
- If  $\gamma \ll 1$  one has the **tunneling** or adiabatic regime.
- If  $\gamma \gg 1$  one has the (frequency dependent) **multi-photon** limit.

- There are two more dimensionless parameters:
  - The ratio between the frequency of the applied field and a typical atomic frequency,  $\omega/\omega_{\text{atom}}$ , whose inverse is  $\sim n$ , the number of photons involved in the process  $\gg 1$ .
  - the ratio of applied electric field amplitude to a typical atomic field strength,  $F/F_{\text{atom}} < 1$ .

- Description in terms of Green's functions

- Schrödinger's differential equation is (we use atomic units)

$$i\frac{\partial|\psi\rangle}{\partial t} = [T + V + V_{\text{ext}}]|\psi\rangle \equiv H|\psi\rangle,$$

where  $T$  is the kinetic energy operator,  $V$  the binding potential, and  $V_{\text{ext}}$  the potential due to the applied field.

- The equation for the Green's function  $G(t, t')$

$$[i\frac{\partial}{\partial t} - H]G(t, t') = i\delta(t - t').$$

- There are two related Green's functions  $V_{\text{ext}}$  only:

$$[i\frac{\partial}{\partial t} - H_0]G_0(t, t') = i\delta(t - t'),$$

$$[i\frac{\partial}{\partial t} - H_{\text{ext}}]G_{\text{ext}}(t, t') = i\delta(t - t').$$

corresponding to the hamiltonians  $H_0 = T + V$  and to  $H_{\text{ext}} = T + V_{\text{ext}}$ , respectively.

- The relationships between  $G$  and  $G_{\text{ext}}$  and between  $G$  and  $G_0$  are given by the following expressions

$$G(t, t') = G_0(t, t') - i \int_{t'}^t dt'' G_0(t, t'') V_{\text{ext}}(t'') G(t'', t'),$$

and

$$G(t, t') = G_{\text{ext}}(t, t') - i \int_{t'}^t dt'' G_{\text{ext}}(t, t'') V(t'') G(t'', t').$$

- We use the second form.
- The transition amplitude can be calculated in terms of  $G$  by

$$T_{fi} = \langle \psi_f(t) | G(t, t_0) | \psi_i(t_0) \rangle .$$

- If we specify the external field as

$$V_{\text{ext}} = F x \cos(\omega t),$$



Then

$$G_{\text{ext}}(x, t; x', t') = \frac{\exp[iS_{cl}(x, t; x', t')]}{\sqrt{2\pi i(t - t')}} ,$$

in terms of the **classical action**  $S_{cl}(x, t; x', t')$  for an electron with current position  $x, t$  and initial position  $x', t'$  which is given by

$$S_{cl}(x, t; x', t') = \frac{1}{2(t - t')} \{ (x - x') - [x_0(t) - x_0(t')] \}^2 \\ - \frac{1}{2} \int_{t'}^t d\tau \dot{x}_0^2(\tau) + x\dot{x}_0(t) - x'\dot{x}_0(t') ,$$

- Combining results, we obtain the integral equation

$$\psi(x, t) = \psi_0(x, t) - \frac{i^{1/2}}{\sqrt{2\pi}} \int_0^t \frac{dt'}{\sqrt{(t - t')}} \int_{-\infty}^{\infty} dx' \\ \exp[iS_{cl}(x, t; x', t')] V(x') \psi(x', t') .$$

- Specialization to the potential  $\delta(x)$

- If now,

$$V(x) = -B \delta(x),$$

then there exists only one bound state, with energy

$$-I_p = -B^2/2,$$

- In this case the integral equation becomes one in time only for  $\chi(t) \equiv \psi(0, t)$ :

$$\chi(t) = \chi_0(t) + \frac{i^{1/2} B}{\sqrt{2\pi}} \int_0^t \frac{dT}{\sqrt{T}} \exp[iS(t, t-T)] \chi(t-T).$$

- Knowing  $\chi(t)$ , one can reconstruct  $\psi(x, t)$ . However, knowledge of  $\chi$  is sufficient to calculate the ionization rate.

- Ionization in a constant Electric field

- In a constant electric field  $F$ , the classical position of an electron is ( $q = -1$  a.u.)

$$x_0 = -\frac{F}{2}t^2.$$

- Then, simply,

$$S(t, t - T) = -\frac{F^2 T^3}{24},$$

and

$$\chi(t) = \chi_0(t) + \frac{i^{1/2} B}{\sqrt{2\pi}} \int_0^t \frac{dT}{\sqrt{T}} \exp\left(-\frac{i F^2}{24} T^3\right) \chi(t - T).$$

- Setting

$$\tilde{\chi}(p) \equiv \int_0^\infty dt \exp(-pT) \chi(t),$$

the transformed integral equation becomes

$$\tilde{\chi}(p) = \frac{\tilde{\chi}_0(p)}{[1 - C(p)]},$$

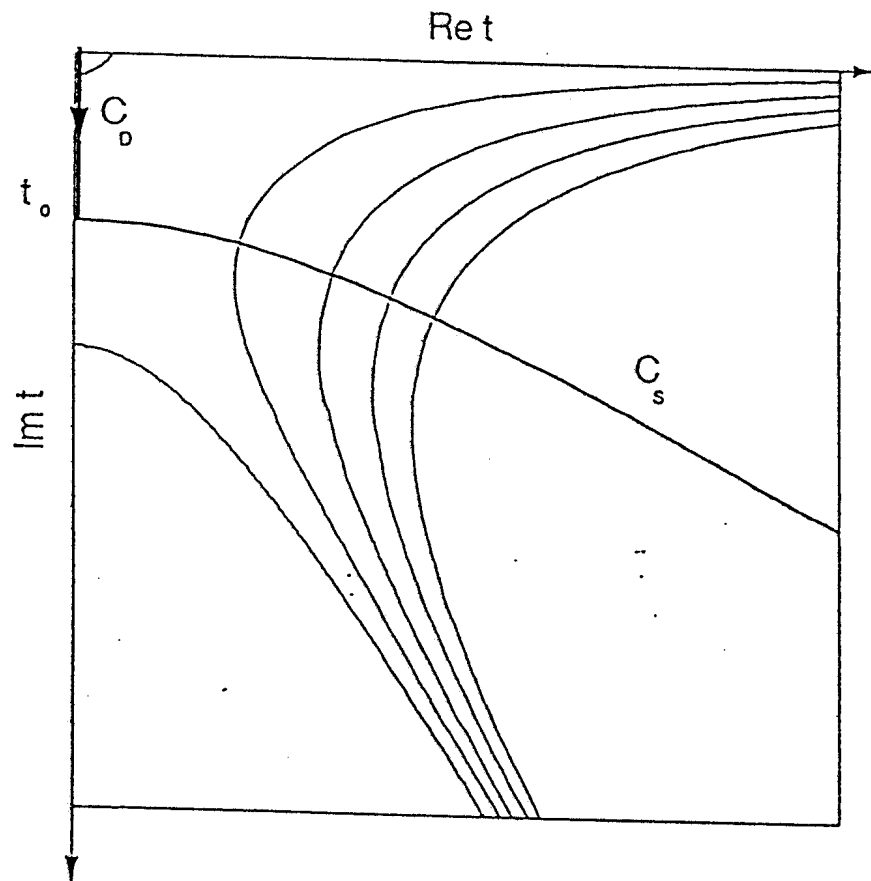
where

$$C(p) = \frac{i^{1/2} B}{\sqrt{2\pi}} \int_0^\infty \frac{dt}{\sqrt{t}} \exp\left(-pt - \frac{i F^2}{24} t^3\right).$$

- With smooth initial conditions, we desire the solution

$$1 - C(p) = 0.$$

- The analysis is similar to Landau's calculation of plasma oscillations.



- Setting  $p = i\epsilon - w/2$ , we write

$$C(p) = C_D\left(\epsilon + i\frac{w}{2}\right) + C_S\left(\epsilon + i\frac{w}{2}\right),$$

where  $\epsilon \simeq \epsilon_0 \equiv B^2/2$

Viewgraph 19

- Ionization in a time-dependent Electric field

- The integral equation is no longer in convolution form.
- We look for a solution of the integral equation in the eikonal form

$$\chi(t) = \exp[i \int_0^t d\tau \epsilon(\tau)],$$

- Substitution into the integral equation, and neglect of the inhomogeneous term, as for the time-independent case yields

$$1 = \frac{i^{1/2} B}{\sqrt{2\pi}} \int_0^t \frac{dT}{\sqrt{T}} \exp\{i[S(t, t-T) - \int_{t-T}^t d\tau \epsilon(\tau)]\}.$$

- Assuming that, for sufficiently weak fields  $F$ ,  $\epsilon$  is nearly real, we obtain, to lowest order

$$\epsilon(t) = \frac{B^2}{2}.$$

from the integral near  $T = 0$ .

- From an expansion of  $C_D$ , we obtain

$$\epsilon = \frac{B^2}{2} \left[ 1 + \frac{5}{4} F^2 / B^6 + \dots \right],$$

$$w = 0.$$

- $w$  is determined by including  $C_S$ ; specifically

$$\frac{w}{2} \frac{\partial C_D}{\partial \epsilon} = -\text{Im } C_S(\epsilon).$$

- Evaluating  $C_S$  and  $\partial C_D / \partial \epsilon$  to lowest order we obtain the ionization rate

$$w = B^2 \exp \left( -\frac{2 B^3}{3 F} \right).$$

- Result for  $w$  is a complicated expression which has the following features:

- Total rate is a sum over the (integer) number  $s$  of photons absorbed where

$$s\omega \geq \frac{B^2}{2} \left(1 + \frac{1}{2\gamma^2}\right)$$

which is the sum of the ionization energy plus the ponderomotive energy acquired by an electron oscillating freely in the applied field.

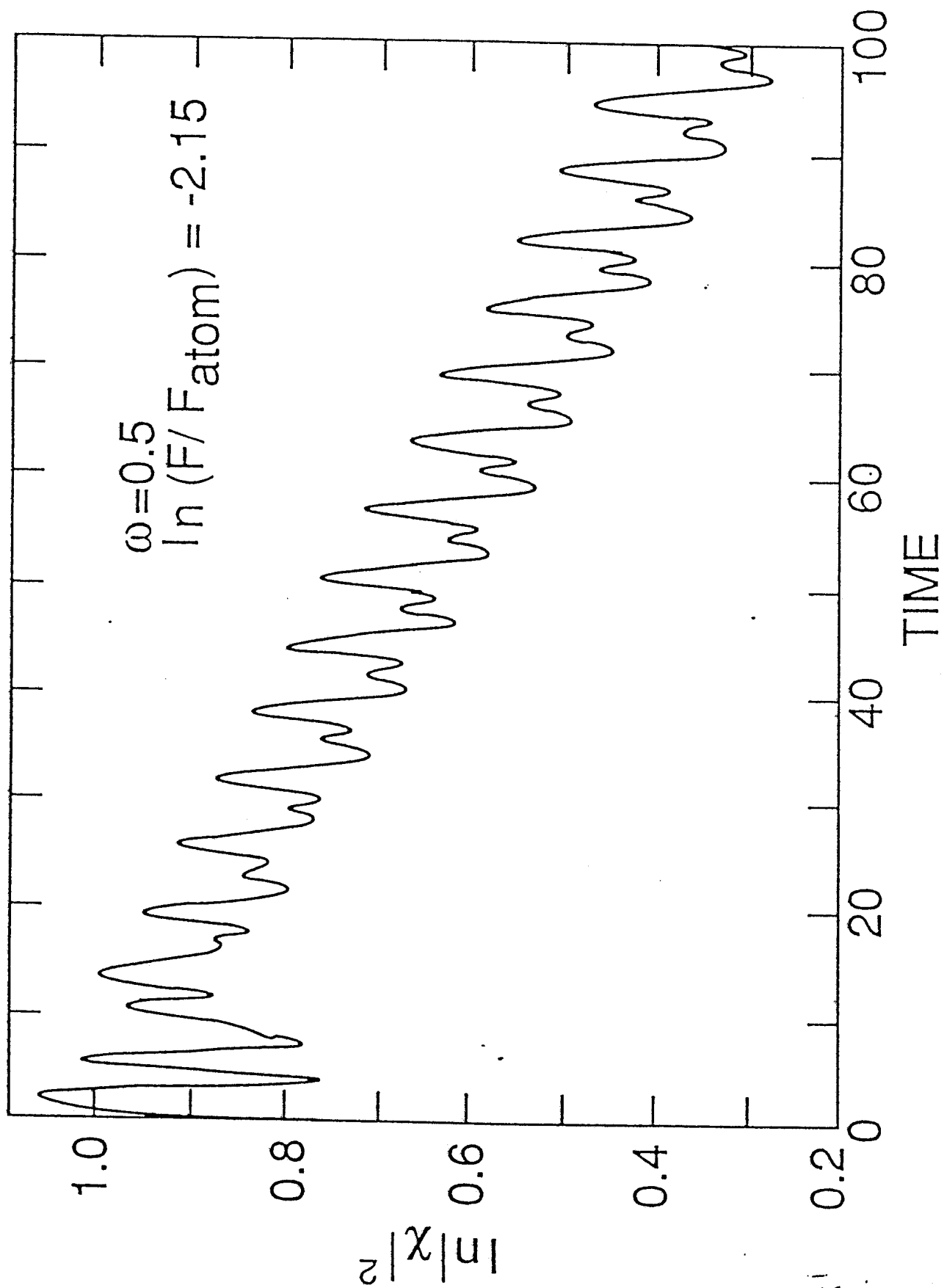
- In the adiabatic limit,  $w$  becomes equal to the period-averaged tunneling result,

$$\begin{aligned} w_{\text{aver}} &= \frac{\omega}{2\pi} \int_0^{2\pi/\omega} w(t) dt, \\ &= \frac{B^2 \omega}{2\pi} \int_0^{2\pi/\omega} \exp\left(-\frac{2}{3} \frac{B^3}{F |\cos(\omega t)|}\right) dt. \end{aligned}$$

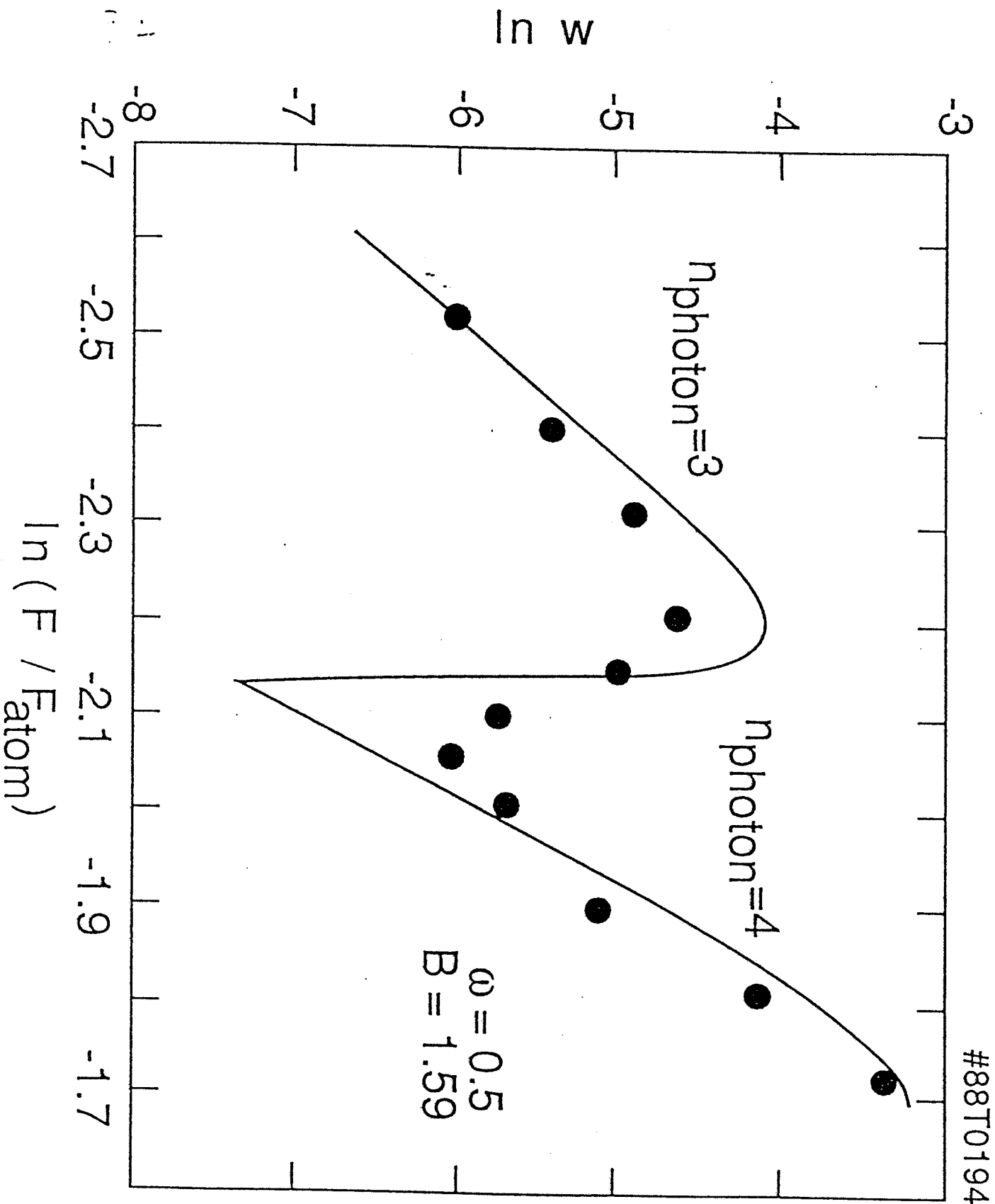
as expected. (Ionization occurs in ‘bursts’ at the instant of maximum field strength.)

- A comparison with a direct numerical solution of the integral equation shows good agreement for even a few-photon process.

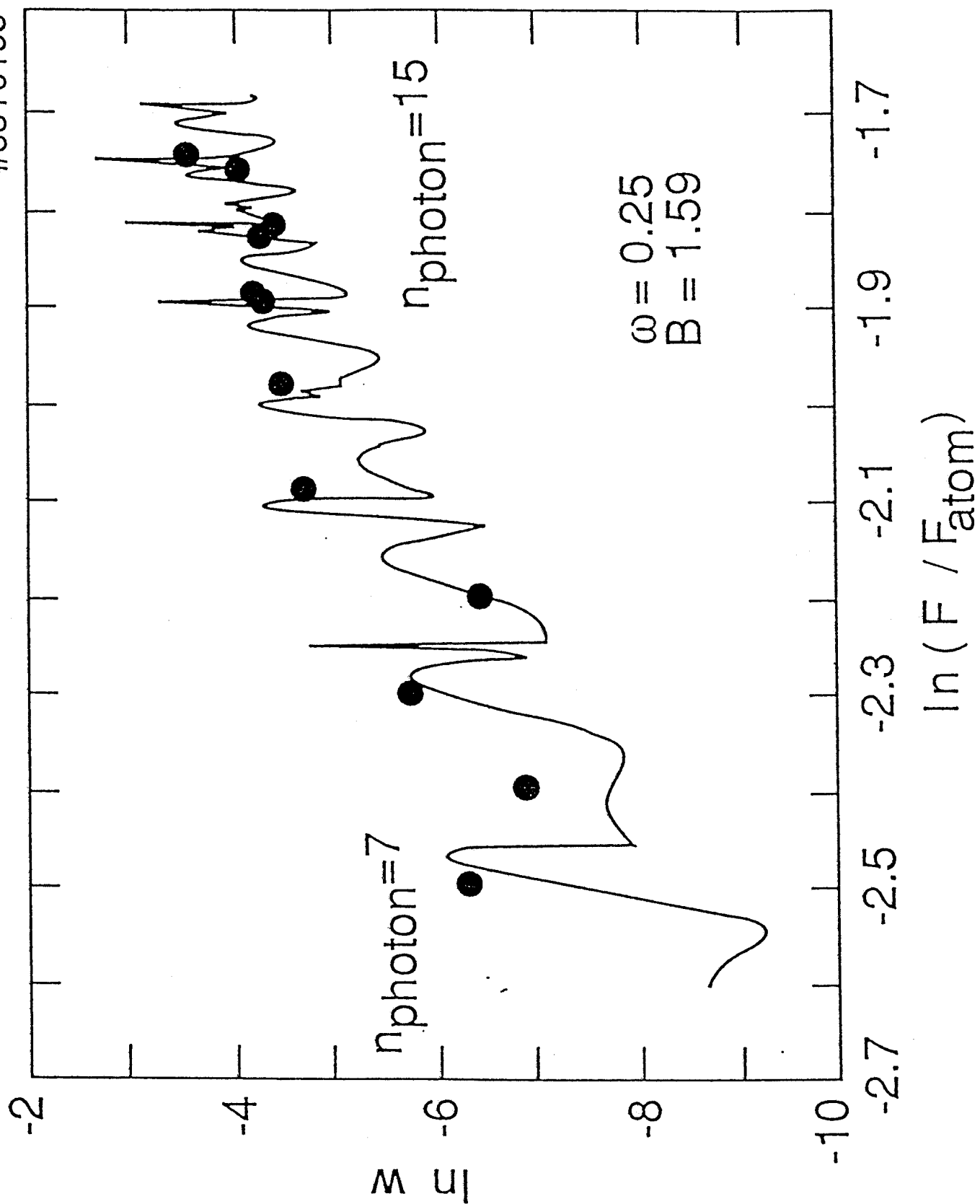
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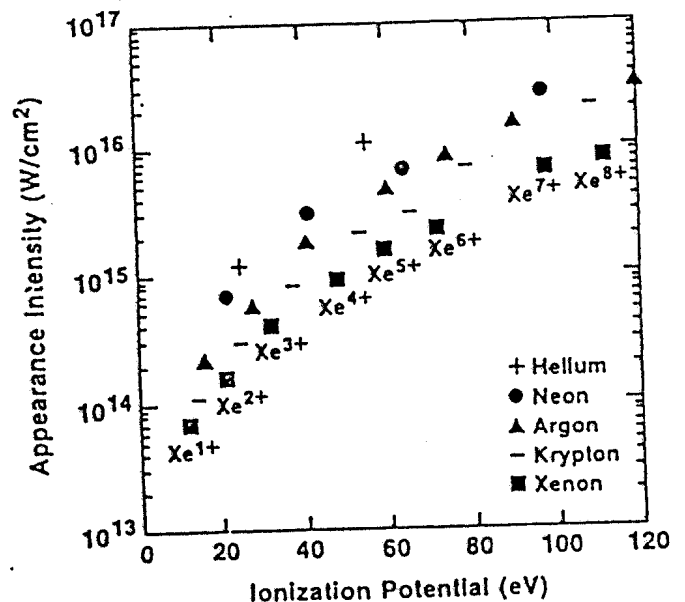
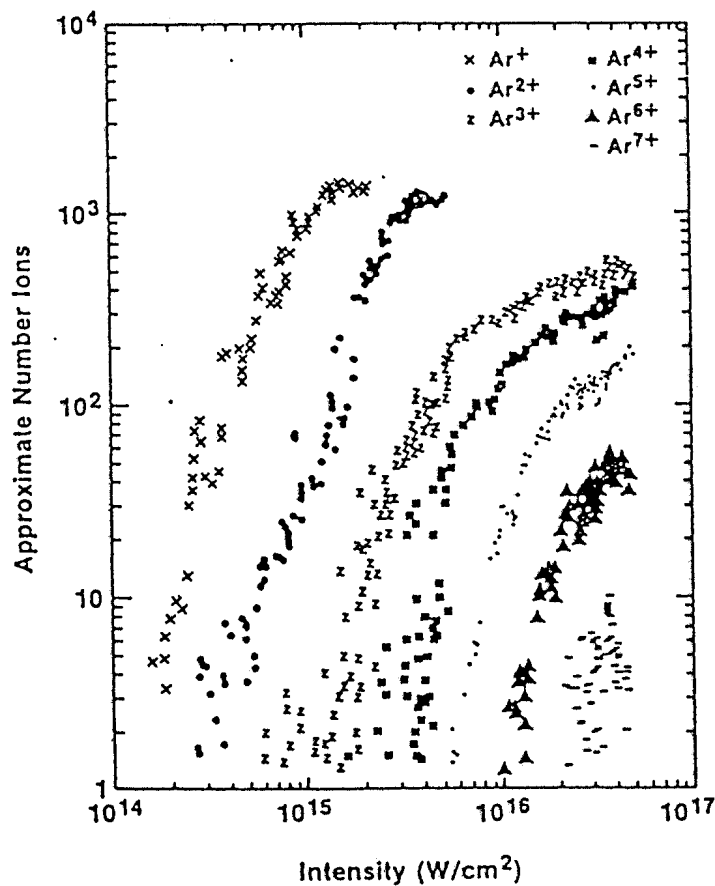


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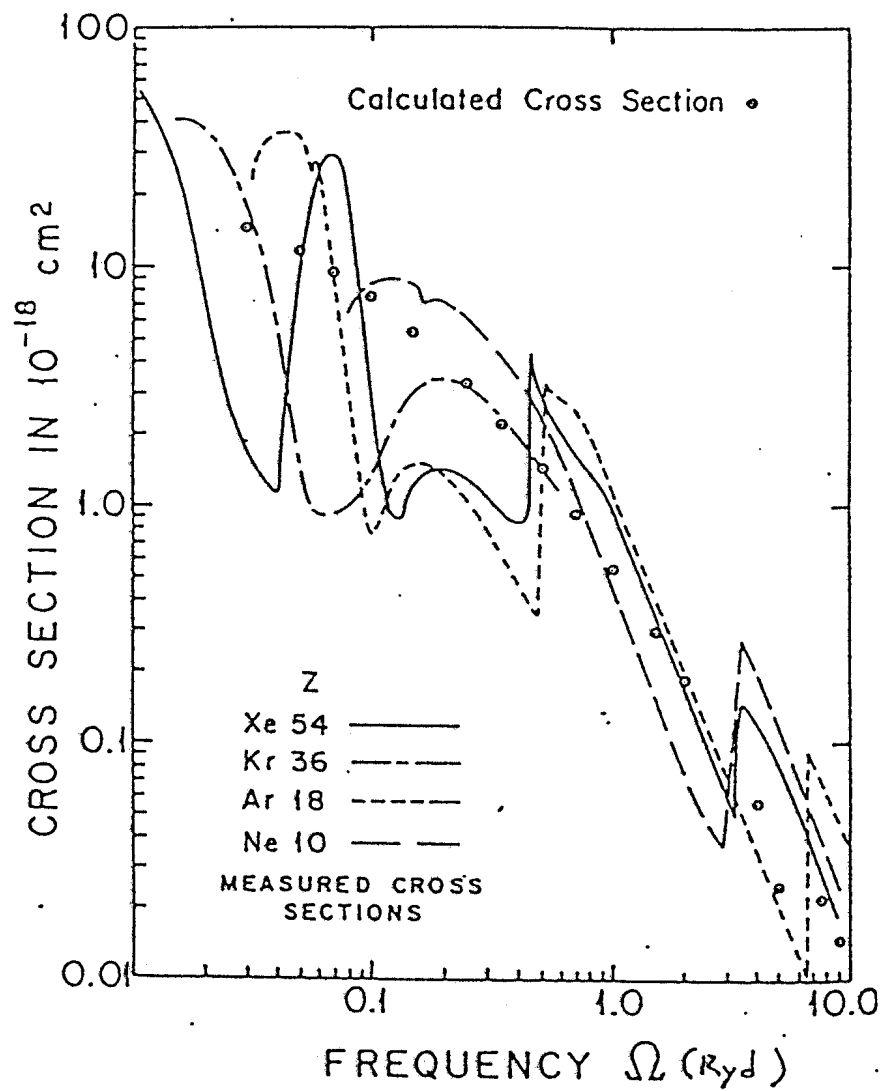
- Non Resonant MPI has generic features

- In the case of nonresonant MPI the threshold intensity for a given ionization stage is observed experimentally to depend most sensitively on the ionization potential of the preceeding stage.



S. Augst, D. Strickland, D. D. Meyerhöfer, S. L. Chin, and J. H. Eberly, Phys. Rev. Letters, 63, 2212 (1989).

- For many-electron ions this lack of dependence on shell structure suggests the possibility of a description by means of a statistical model, such as the Thomas-Fermi model:
  - Semiclassical model which accounts for screening properties of many-electron atoms.
  - Large  $Z$  expansion: appropriate for many electron systems.
  - However, it misses shell structure and tunneling.
  - Past successes include:
    - \* Calculation of atomic photoabsorption cross-section.
    - \* Computation of atomic properties such as volume vs  $Z$  and radial charge density.



J. A. Ball, J. A. Wheeler, and E. L. Fireman, Rev. Modern Physics, 45, 333 (1973).

- The TF model follows systematically from a large  $Z$  expansion

Many-electron Schrodinger equation  
neglect dynamical correlation  $\sim Z^{-4/3}$



Hartree-Fock approach (independent particles)  
neglect exchange  $\sim Z^{-2/3}$



Hartree equations  
neglect quantum effects  $\sim Z^{-2/3}$



Thomas Fermi (keep terms up to  $Z^{-1/3}$ )

- The TF model is relatively easy to solve
- The model describes a zero-temperature, electrostatically confined (by the nuclear field) Fermi gas of electrons interacting through the self-consistent mean field.
- The dependence of electron density  $n_e$  on potential  $\phi$  is computed by:

– Dividing phase space into cells where

$$\# \text{ of cells} = \frac{\Delta p \Delta x}{(2\pi)^3} = \frac{1}{2} \# \text{ of electrons}.$$

– Filling phase space completely for momenta

$$p \leq p_{\max} \equiv [2(\phi - \epsilon)]^{1/2},$$

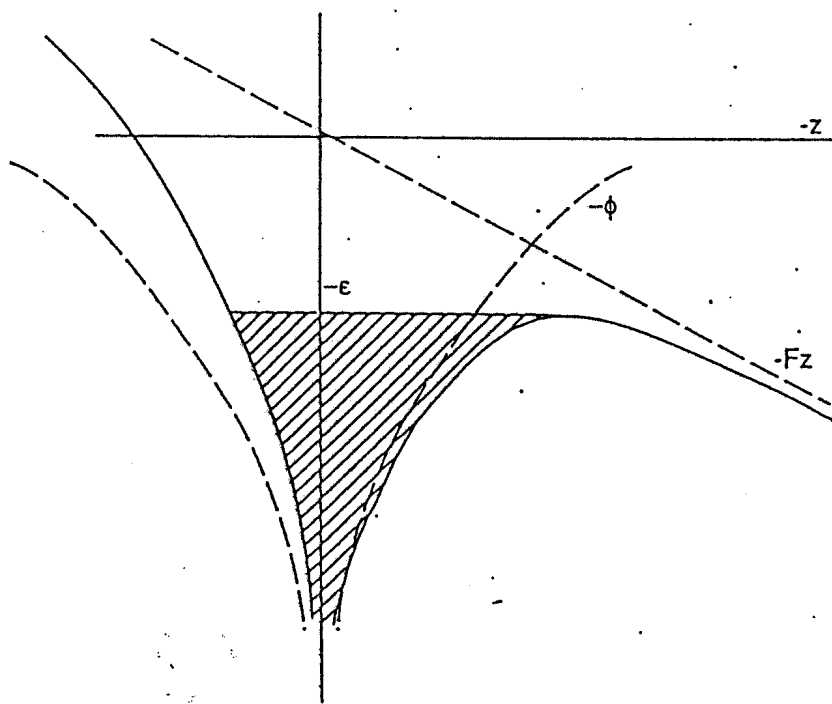
where  $\epsilon$  is the Fermi energy.

- The electron density is then determined from the local potential  $\phi$  and from  $\epsilon$

$$n_e = 4\pi \int_0^{p_{\max}} dp p^2 \frac{1}{4\pi^3} = \frac{p_{\max}^3}{3\pi^2} = \frac{2^{3/2}}{3\pi^2} (\phi - \epsilon)^{3/2}$$

When  $\phi < \epsilon$ ,  $n_e \equiv 0$ .

- We include a static electric field  $\vec{F}$  in the  $\hat{z}$  direction by making the replacement  $\phi \Rightarrow \phi + \vec{F} \cdot \vec{r}$ 
  - We associate  $F$  with the peak laser electric field (assuming linear polarization)  $E(t) = F \cos(\omega_{\text{laser}} t)$ , since  $\omega_{\text{laser}} \ll \omega_{\text{atomic}}$ .
  - No tunneling. (Shown to be negligible beyond the first ionization stage for short pulse experiments considered.)
  - “Tipping bucket model”



- $\epsilon$  is now the energy at the separatrix when  $F \neq 0$  and is the ionization potential when  $F = 0$



- Poisson's equation, together with the boundary conditions

$$\begin{aligned} r\phi(r, \mu) &\rightarrow Z \quad \text{as } r \rightarrow 0, \\ r\phi(r, \mu) &\rightarrow Z - Q + Fr^2\mu \quad \text{as } r \rightarrow \infty, \end{aligned}$$

where

$$Q \equiv \int d^3r n_e,$$

completes the description.

- A numerical solution yields, besides  $Q$ , the dipole moment

$$\langle z \rangle \equiv Q^{-1} \int d^3r z n_e,$$

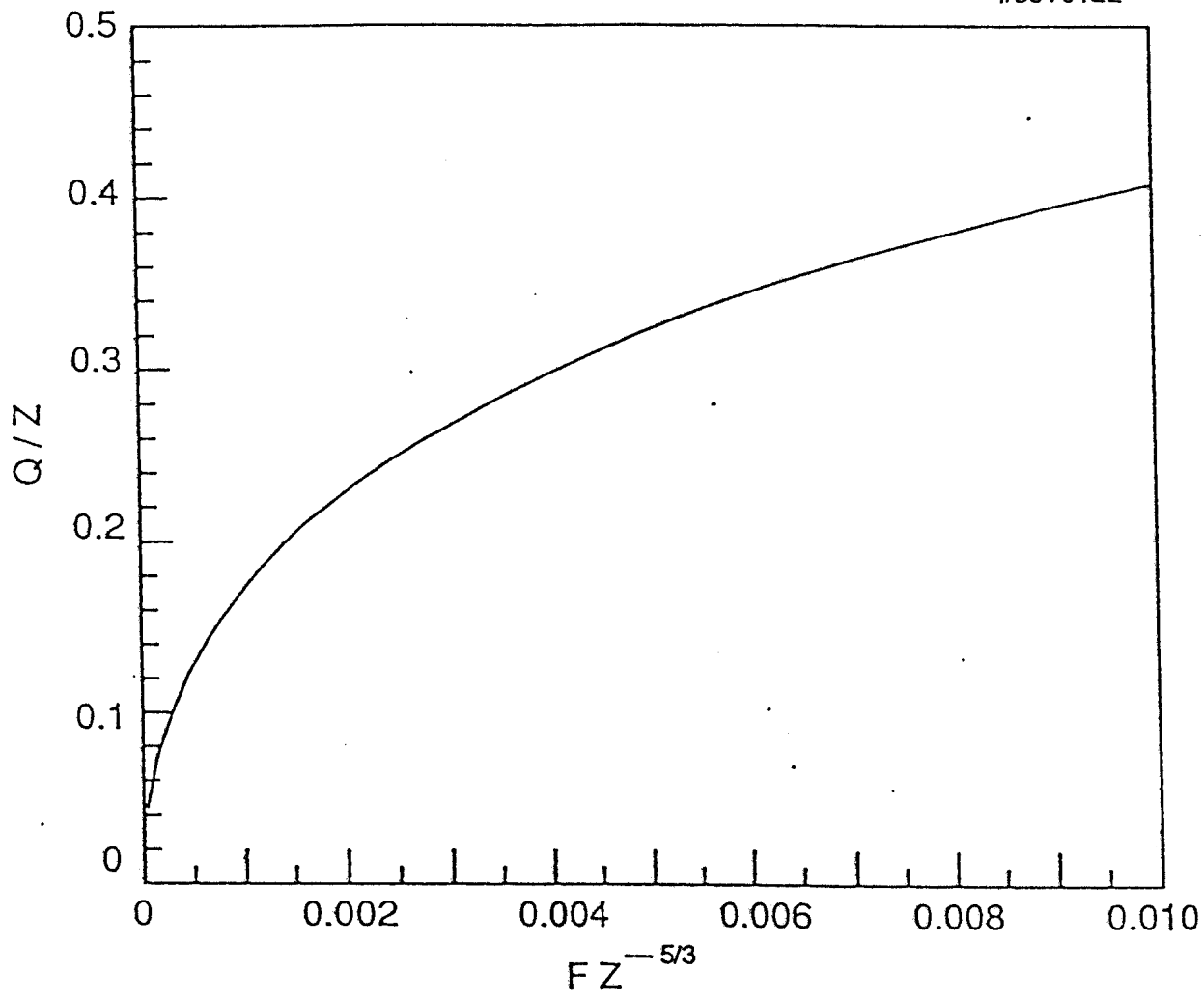
vs  $F$ .

- To compare with experiment, we define the ionization potential as

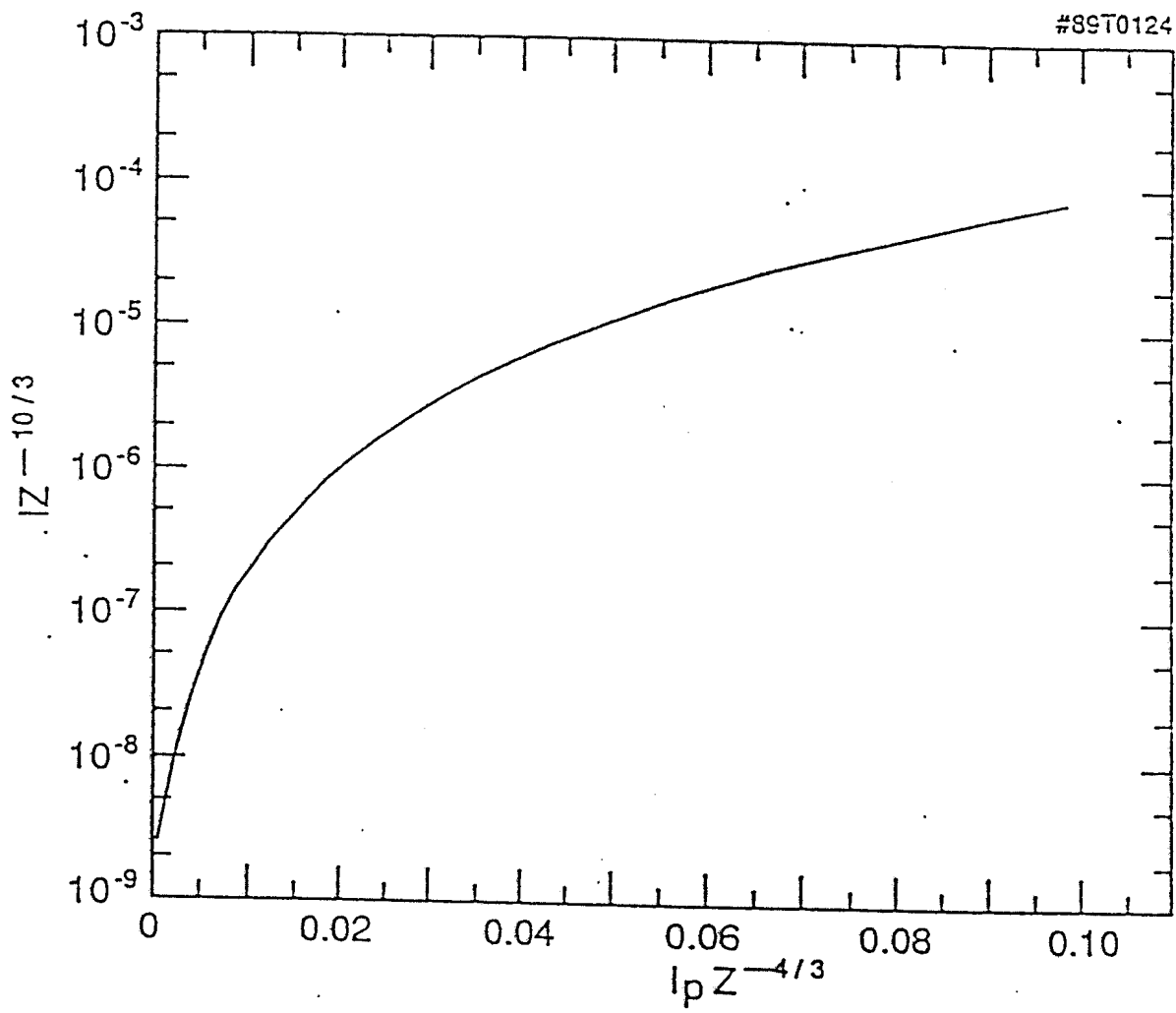
$$I_p \equiv \epsilon|_{F=0},$$

- In a separate sequence solve for  $I_p$  as a function of  $Q|_{F=0}$ .
- The model yields universal curves of  $Q/Z$ ,  $\langle z \rangle Z^{1/3}$  vs  $FZ^{-5/3}$  and of  $Q/Z$  vs  $I_p Z^{-4/3}$ .
- Together, they yield  $F(I_p)$ .
- There are no adjustable parameters.

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Viewgraph 33



Viewgraph 34

